

DOSE ESTIMATION BY BICASSAY FOR POPULATION INVOLVED IN AN
ACCIDENT WITH PLUTONIUM RELEASE

Emilio Iranzo, Asunción Espinosa and C.Emma Iranzo.
Instituto de Protección Radiológica y Medio Ambiente
C.I.E.N.A.T. SPAIN

SECOND CONFERENCE ON RADIATION PROTECTION AND DOSIMETRY

Orlando, Florida. 31 October - 4 November 1988

DOSE ESTIMATION BY BICASSAY FOR POPULATION INVOLVED IN AN
ACCIDENT WITH PLUTONIUM RELEASE

Emilio Iranzo, Asunción Espinosa and C.Emma Iranzo.
Instituto de Protección Radiológica y Medio Ambiente
C.I.E.N.A.T. SPAIN

SECOND CONFERENCE ON RADIATION PROTECTION AND DOSIMETRY

Orlando, Florida. 31 October - 4 November 1988

DOSE ESTIMATION BY BIOASSAY FOR POPULATION INVOLVED IN AN
ACCIDENT WITH PLUTONIUM RELEASE

Emilio Iranzo, Asunción Espinosa and Emma Iranzo
Instituto de Protección Radiológica y Medio Ambiente
C.I.E.M.A.T. Spain

1. INTRODUCTION

On January 17, 1966 while flying over Palomares (Almería) in the Southeast of Spain two United States Air Force Planes collided while on a mid-air refuelling operation. One of the planes carrying four thermonuclear weapons, three of which, one intact, were found on land in or near Palomares, 24 hours after the aircraft wreckage. The fourth thermonuclear weapon was found in the Mediterranean Sea on April 7. The parachutes of two of the bombs did not deploy, resulting in the detonation of their conventional explosives and release of their fissile material upon impact. Partial ignition of the fissile material formed a contaminating cloud which covered approximately 226 hectares (558 acres) of uncultivated, farm and urban land as shown in Figure 1.

One of the bombs landed about 1 mile to the West of Palomares (impact point number 2) and the plutonium-bearing dust cloud was carried by 30-knot West winds, over non-cultivated terrain, irrigated farming fields and the northern edge of the village. The cloud from the other fragmented bomb, which landed

in the East edge of the village, travelled away from the village but across farmed areas.

Visible bomb fragments and pieces of the aircraft were collected and surface alpha contamination measured by PAC-15 alpha detectors.

Most of the houses in the village were not contaminated. Some were contaminated at low levels and washed with high-pressure water and detergents. Residual contamination was removed by hand and some houses were white-washed.

Figure 1 shows the various levels of the originated surface alpha contamination and the extension corresponding to each level of alpha contamination.

The houses were located in the area with the least level of contamination, and a great many of them in areas that were not contaminated at all. Since the day of the accident was a holiday, very few people were working in the fields, but some hours later most of the population went out over the area to observe the aftermath of the accident. On 19 January 1966, people were forbidden from entering the contaminated areas and were requested to stay in their homes and the urban area where the levels of alpha contamination in the air were lower than 3.7 mBq/m^3 (10^{-13} Ci/m^3).

In order to acquire a basic idea of the possibility of contamination of the residents of Palomares, external contamination was measured on most of them just after the accident. The highest contamination level was 33 Bq/100 cm^2 (0.9 nCi/100 cm^2).

Early urine samples from people and military personnel to determine internal contamination were often contaminated during sample collection.

Remedial actions taken according to the knowledge of the time, so there would not be any unacceptable risk in the short and long term for inhabitants and people farming in the area with transuranides residual contamination, are shown in Table 1.

Once remedial actions were completed and radioactive wastes were removed, an experimental radiological surveillance programme was established in order to study short, medium and long-term effects on people and the environment.

The main objectives of the surveillance programme with regard to the people who inhabit and cultivate the transuranide contaminated area were as follows:

- To determine the level of internal contamination of the inhabitants of the area during the time immediately following the accident and the emergency phase.

- To determine the risk of internal contamination for the people who inhabit and cultivate the area.

In relation to these objectives, this report deals specifically with the plutonium measurements in urine carried out over a period of 22 years on 714 residents of Palomares, the evaluations on internal contamination due to inhaling which have been deduced on the basis of these measurements, and the estimations of the committed effective dose equivalent that have been carried out on 55 inhabitants considered as being internally contaminated.

In 1966, urine samples of inhabitants were taken in Palomares at three different dates. Measurements of Pu in urine were made at the JEN in Madrid. Sampling and measurements of Pu in urine and lungs of Palomares inhabitants were started at the JEN in Madrid in 1967 and have continued uninterrupted since

1975. Every year 150 residents are subject to medical examinations in addition to the measurements to detect Pu.

2. METHODOLOGY FOR ANALYSIS AND DOSE CALCULATION

2.1 Plutonium analysis

The analyses to determine the plutonium excreted in urine are carried out in 24-hour samples.

The radioanalytical technique used to determine the concentration of ^{239}Pu and ^{240}Pu is basically the same as that used in Los Alamos National Laboratory, USA (Ev66, Ga83). It includes the acidification of the sample with concentrated HNO_3 , the coprecipitation of the plutonium with earthy-alkaline phosphates, the wet incineration of the precipitate with concentrated HNO_3 at 300°C , the separation of the Pu in solution from the Am and U by means of Dowex AG 1 x 2 anionic resin and washing with 8N HNO_3 , the elimination of the Th by means of concentrated HCl , the elution of the Pu fixed in the resin with 0.5N HCl in the presence of hydroxylammonium-chloride, and the electrodeposition of the Pu on stainless-steel planchets in a medium of 1N HCl with 4% ammonium oxalate.

The ^{239}Pu and ^{240}Pu in the planchets are measured by alpha spectrometry with Si barrier detectors, having an active area of 300 mm^2 , a resolution of 21 keV and a dead time of 0.5 microseconds. The counting efficiency of the equipment is in the order of 30%.

The chemical yield of each analysis is measured by addition of ^{242}Pu as a tracer. It is about 75%.

2.2 Dose calculation

The criteria and techniques used for estimating the committed effective dose equivalent in the case of those persons considered as being internally contaminated by plutonium on account of the results of the excretion of $^{239}\text{Pu} + ^{240}\text{Pu}$ in the urine, have been as follows:

- The intake is due to acute inhalation of particles of $1\ \mu\text{m}$ (AMAD), class Y plutonium at the time of the accident or during the days immediately afterwards, for the people who were in Palomares when it happened.

Chronic inhalation was rejected upon discovering that the positive values of excretion in urine that were obtained would require the concentration of plutonium activity in the surrounding air to be constantly very much higher than the plutonium-concentration values measured in the area (Ir 87) during this 22 year period. The average concentration in air measured in the urban zone was around $5\ \mu\text{Bq}/\text{m}^3$ ($0.16\ \text{fCi}/\text{m}^3$) and about ten times higher in the area with the greatest level of concentration.

In order to fix the date of the probable intake due to inhalation in the case of those people who were not in the area or who had not been born in 1966, account has been taken of the date on which they came into the area, the type of work and habitat, the years in which, owing to particular circumstances, higher concentration of Pu in the air were measured in the zone, and the correlation of the Pu values in urine with the excretion curve.

- Determining the activity of $^{239}\text{Pu} + ^{240}\text{Pu}$ intake through inhalation on the basis of the values of excretion in urine on the date of the sample, by means

of the method developed by K.Eckerman(Ec84) in the Oak Ridge National Laboratory. This method is based upon the metabolic intake and pulmonary distribution model of the ICRP and on the urinary excretion model of Langham modified by W. Moss (Moss 83) in Los Alamos National Laboratory. In the case of those people who have had Pu measurements in urine higher than the MDA, it was considered that their MDA values were positive and with a value equal to the limit.

The equation which, in the model referred to, describes the time-based urinary excretion of plutonium owing to a particular plutonium activity intake through inhalation, is as follows:

$$U_e(t) = \int_0^t A(\tau) \cdot Y(t-\tau) d\tau$$

if:

- U_e Urinary excretion.
- $A(\tau)$ Function describing the passage into the bloodstream of the activity intaken through inhalation.
- $Y(t-\tau)$ Function describing the urinary excretion of the plutonium in the system.

For experimental studies, estimations have also been made which are based on the same method but with the urinary excretion curve developed by Jones(Jo85).

- Calculation of the committed effective dose equivalent by means of the Sv/Bq conversion factors recommended

by the ICRP (ICRP79) in order to calculate the weighted committed dose equivalent for the most important organs from the point of view of their contribution to the committed effective dose equivalent (red marrow, lungs, bone surface, liver).

For experimental purposes, estimations have also been made of the committed effective dose equivalent for 70 years, using for persons of different ages the Sv/Bq conversion factors established by Kaul and his collaborators (He85).

3. RESULT AND DISCUSSION

The results of the measurements of ^{239}Pu and ^{240}Pu in the urine samples taken over a period of 22 years from those Palomares residents who have been checked at least once, as well as the estimations of the committed effective dose equivalents which, according to the results, have been considered as being contaminated, are discussed below.

3.1 Urine concentration of Plutonium

In 1966, urine samples were taken in Palomares from 59 people who were considered to have been exposed to the greatest risk of internal contamination, on account of where they were at the time of the accident and the measurements of external alpha contamination that were taken. Samples were collected from each person on three occasions between 6/6/66 and 10/8/66. Pu in urine above MDA was measured in 57 people (96.6%) in one or more samples.

The values of the ^{111}Pu measurements ^(above MDA) that were carried out fell within the range of 0.74-17908 mBq/day. The average value of the measurements was 244 ± 1734 mBq/day. This, together with the fact that those people who showed higher values also

had values that were around the MDA, suggested to us the possibility of external contamination of the urine samples, and it was thus decided that the latter should be taken outside the Palomares area.

Since 1967, those people from Palomares who have been undergoing annual examinations have been travelling to Madrid in order to have urine samples taken, to have direct lung contamination measurements (Chest Counter and Whole Body Counter) and to be given a medical examination. The chest measurements using the most sophisticated detectors for the time indicate no Pu above the minimum detectable activity (MDA) of 314 Bq (22 nCi).

In 1967, the same 59 people who had been looked at in 1966 were examined in Madrid, and 24-hours urine samples were taken from them for 3 consecutive days. Pu in urine above MDA was measured in 23 people (39%) only. The results of the Pu measurements that were carried out fell into the range of 0.37 - 17.4 mBq/day. The average value of the measurements was 2.6 ± 3.49 mBq/day. The study of the Pu urine excretion results as a whole and those corresponding to each individual person shows that could be a factor 4 of variability between samples taken in different days of short periods of time.

As from 1968, the procedure has been to take one 24-hour sample from all the people who have been examined. The results higher than the MDA that have been measured since then are in the range of 0.74 - 51.8 mBq/day with an average value of 6.4 ± 9.5 mBq/day.

As was pointed out before, up until 1st January 1988, 714 people had been examined, about 93% of the residents and 1815 measurements of Pu in urine had been carried out. 425 people out of those who were examined were in Palomares at the time of the accident and 229 were not there, either because they were born at

a later date or because they had fixed their residence there afterwards. Table 1 provides a breakdown of the people in terms of their sex and the number of analyses carried out on them.

124 people had shown, at least once, values of Pu in urine higher than the MDA [0.37 MBq/day (0.01 pCi/day)]. Of these, 100 were there at the time of the accident and 24 came into the area at a later date.


On the 1815 analyses of Pu in urine carried out, 234 had given values higher than the MDA. The distribution of these values, as regards the people affected and in relation to the number of positives and the total number of tests made on the corresponding people, is shown in Table 2.

In accordance with the results obtained from the plutonium in urine tests made on the 714 Palomares residents who had been examined in one occasion or more, these people were classified into the following three groups:

Group 1: With results lower than the MDA - 590 people.

Group 2: With results higher than the MDA in the samples taken in Palomares (1966) and lower than the MDA in those taken in Madrid - 29 people.

Group 3: With some result higher than the MDA in the samples taken in Madrid - 95 people.



3.2 Dose Estimation.

The estimation of the committed effective dose equivalent was carried out on the 55 people who were considered really to have suffered internal contamination, by making a rigorous

study, for each of them, of the Pu values in urine, the possibility of external contamination of the sample and its greater or lesser correlation with the model developed by K.Echerman on the basis of ICRP pulmonary model and W.Langham's excretion curve modified by W.Moss. All of these people belong to Group 3.

In order to study the correlation between the Pu values in urine and Echerman's model, a far-reaching study was made on the 30 people who had shown values higher than the MDA in three or more samples. All of them had been given at least 3 tests during the 22 years following the accident, and 60% of them had a minimum of 3. The adjustment of the different values of Pu urine excretion of each person to the curve relating them with the acute inhalation intake on a particular date, led us to the conclusion that, by calculating the intake starting from each one of the values of Pu in urine for the date of the sample, and determining the average value of the intakes that were calculated, we could obtain the most suitable estimation of the intake due to acute inhalation in the case of each person.

Consequently, by following this procedure, calculations were made for the intake of Pu due to acute inhalation for the 45 people, 24 men and 21 women, who were in Palomares when the accident occurred. On the basis of these intakes, and using the Sv/Bq conversion factors of the ICRP (ICRP79), estimations were made of the committed effective dose equivalents corresponding to these people.

In the case of the 10 people, 5 men and 5 women, who were not in the area when the accident took place, it is difficult due to the shape of the curve for the excretion of Pu in urine, to determine the date of intake on the basis of the factors indicated in the methodology. In fact, the doses were estimated in terms of the date of intake considered to be the most likely.

The values of the committed effective dose equivalent estimated for the 55 people in question fall between 20 and 200 mSv (2-20 rem). Table 3 gives a breakdown of these people in terms of intervals between doses.

The study made into the doses calculated for these 55 people reveals a normal logarithmic correlation. Having calculated Henry's straight line, and with the established logarithmic types having been represented, it can be seen that all of them are within the 95% confidence determined by the calculation of Leveau's abacus (Fig. 2.). This fact confirms that the criterion followed for calculating the possible intake by using the averages of the intakes calculated for each excretion is right enough.

One of the person considered internally contaminated at the time of the accident was less than 1 year old, five people were between 5 and 10 years old, and four people were between 10 and 15 years old. The estimation of the 70 years committed effective dose equivalent ($S_{E,70}$) as a function of the age at the time of the acute inhalation intake (He85) has given doses higher in a factor of 3.7, 1.7 and 1.4 respectively to those calculated using the Sv/Bq ratios of ICRP (ICRP79). Accordingly, the $S_{E,70}$ for the child less than 1 year old is 242 mSv (24.2 rem). The $S_{E,70}$ for the others 9 people are in the range of 49-157 mSv (4.9-15.7 rem).

4. CONCLUSIONS

As a result of the surveillance program drawn up for the people of Palomares in order to determine the magnitude of the risk for the population and the application of the Eckerman method to the Pu urine excretion values in the 714 people tested during a period of 22 years, the following conclusions have been reached:

1. As a result of the accident, part of the population, at least 45 people, suffered internal contamination due to inhalation. The committed effective dose equivalents received by this group are between 20 and 200 mSv (2-20 rem). The rest of the population that might have been contaminated would have received a committed effective dose equivalent of below 20 mSv (2 rem).
2. It seems to follow that a group of 10 people may have been internally contaminated on account of their habitat and their participation in the agricultural activities of the area, without it really being possible to establish the time in which the intake could have taken place. Nevertheless, as a result of the suppositions that have been made, it has been estimated that the committed effective dose equivalent that would be received is between 35 and 180 mSv (3.5 - 18 rem). More urine test are necessary to confirm the suppositions and dose estimations.
3. The 70 years committed effective dose equivalent ($S_{E,70}$) for those people less than 15 years old, who were internally contaminated at the time of the accident, is only higher than 200 mSv (20 rem) for 1 person (242 mSv). The $S_{E,70}$ for the others 9 people are in the range of 49-157 mSv (4.9 - 15.7 rem).
4. The assumption made by Legget (Le85) about a periodical remobilization of Pu deposited initially on bone surfaces, could be confirmed at least in three cases, 1 man and 2 ladies. An increase of Pu urine excretion have been measured about 20 years after the accident in these 3 cases.

5. The human metabolic model for plutonium still shows important uncertainties. The estimation of systemic burden of plutonium from urine analyses has experienced a notable development with the proposal made by Jones (Jo85) and Moss (Mo83) for the Pu urine excretion model and the physiological model for Pu developed by Leggett (Le85). On the basis of the experimental values that are being obtained, we shall try to make our own contribution to finding a solution. For the time being, however, the impossibility of obtaining tissue samples from the autopsies of deceased persons is proving to be a big obstacle.

ACKNOWLEDGMENTS

We wish to express our appreciation to the U.S. Department of Energy and its predecessor agencies for helping carrying out this work, as well as to Dra. M.D.Lara and the technicians of the CIEMAT, Carmen Barros, Mariano Casado, José M. Montero and Francisco Moreno. We also wish to thank C.R.Richmond and K.F.Eckerman, Oak Ridge National Laboratory, and W.D.Moss, Los Alamos National Laboratory, for advice and assistance.

REFERENCES

- Ec84 Exxerman K.F., 1984, private communication. Oak Ridge National Laboratory.
- Ev66 Evan Campbell and W.D.Moss. 1966; Health Physics, 11, 737-742.
- Ga83 Gautier, M.A., copiler, 1983, Manual of Analytical Methods for Radiobioassay. LA-9763-M.
- He85 Henrichs K., Elssasser U., Schotola Ch. and Kaul A. 1985. "Dosisfaktoren für inhalation oder Ingestion von Radionuclideverbindungen" in: ISH-HEFT 63, 78, 79, 80 and 81. (Institut für Strahlenhygiene des Bundesgesundheitsamtes, Neuherberg b/München)
- ICRP79 International Commission on Radiological Protection, 1979. "Limits for intakes of radionuclides by workers". ICRP Publication 30, Supplement to Part 1 (Oxford: Pergamon Press).
- Ir87 Iranzo E, Salvador S and Iranzo C.E. 1987. "Air concentrations of ^{239}Pu and ^{240}Pu and potential radiation doses to persons living near Pu-contaminated areas in Palomares, Spain" Health Physics. 52, 453-461.
- Jo85 Jones S.R., 1985. "Derivation and validation of a urinary excretion function for plutonium applicable over tens of years post uptake" Radiation Protection Dosimetry, 11, 19-27

- Le85 Leggett, R.W. "A model of the retention, translocation and excretion of systemic Pu", Health Physics, 49, 1115-1138.
- Mo83 Moss W.D., Tietjen G.L., Gautier M.A. and McLeod M.J., 1983. "A review of the human plutonium injection studies", in Proc. of the 29th Annual Conf. on Bioassay, Analytical and Environmental Chemistry, 12-13 Oct. 1983.

Figure 1. PALOMARES AREA: ORIGINAL CONTAMINATION LEVELS

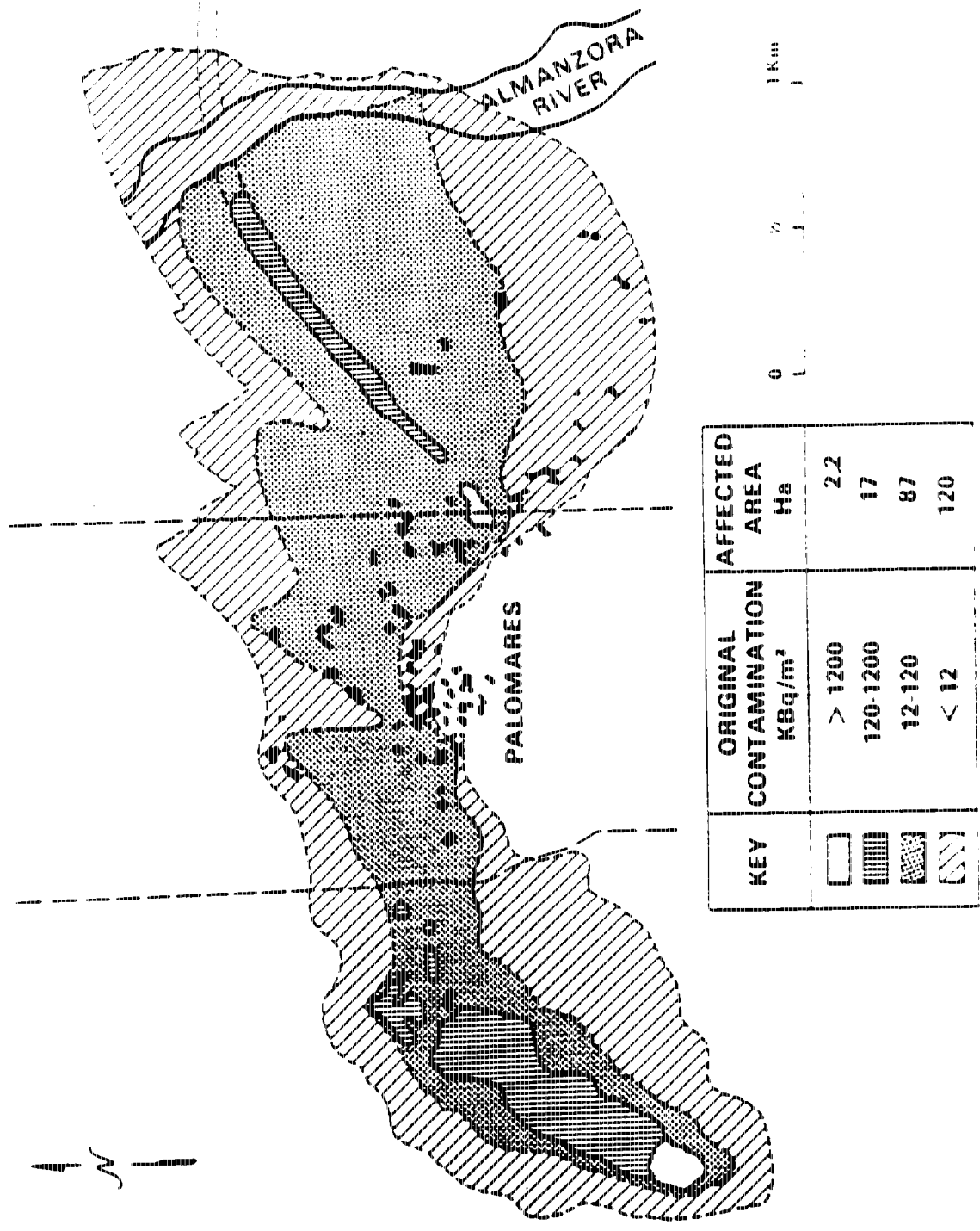


Figure 2. Pu URINE EXCRETION ESTIMATES FOR ACUTE INHALATION INTAKE (AMAD = 1 μ m)

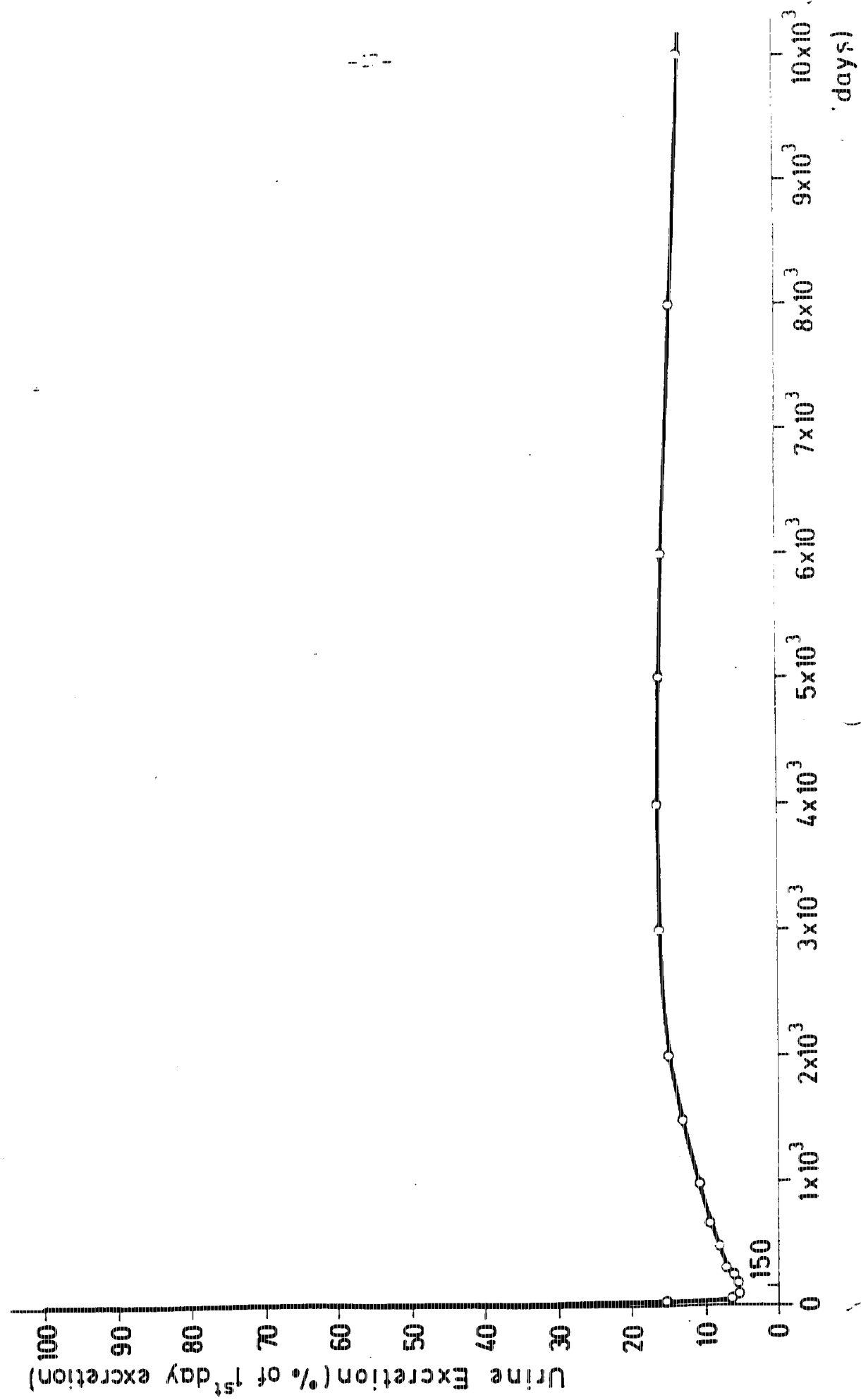
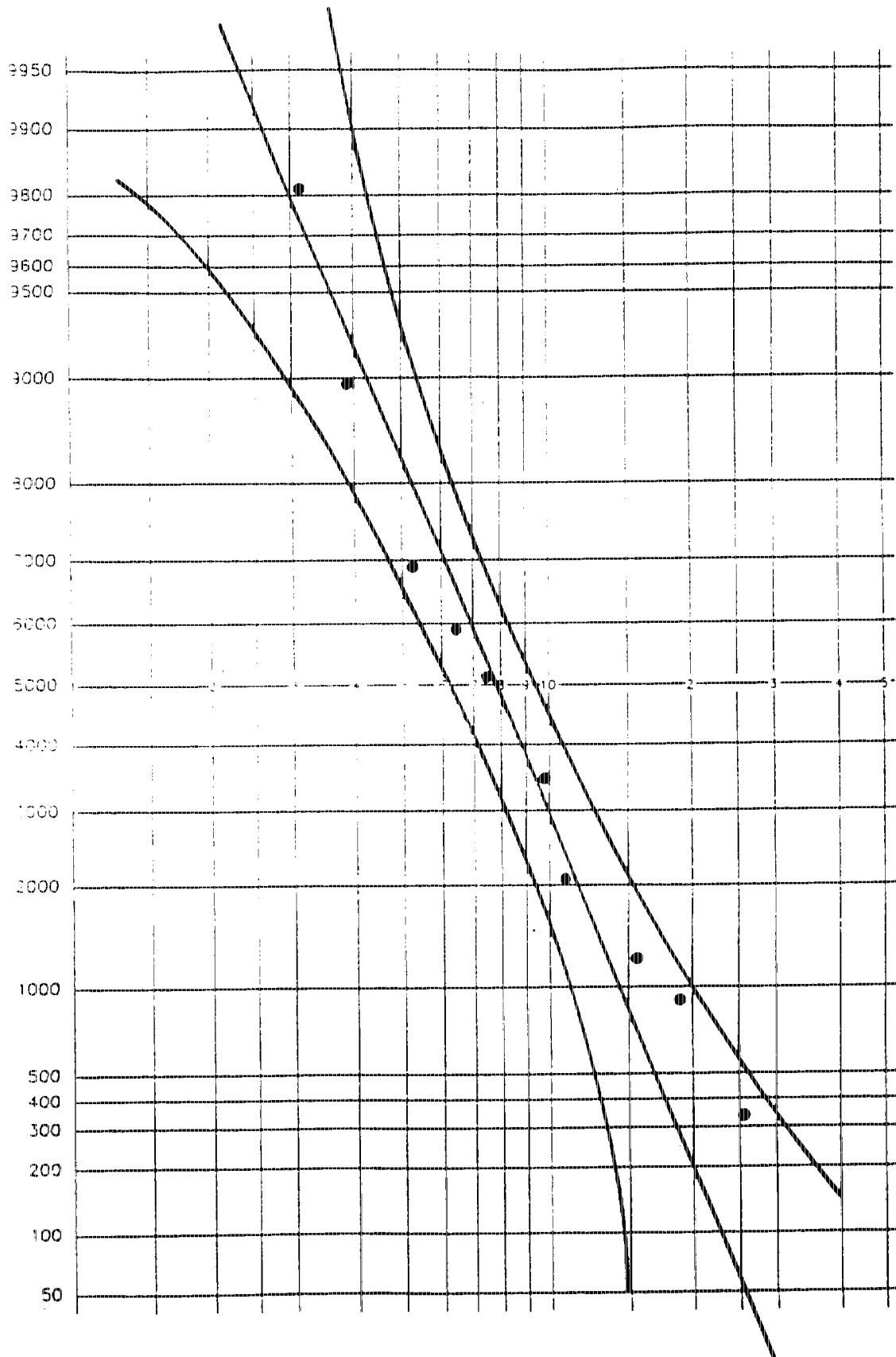


Figure 3. DISTRIBUTION OF ESTIMATED $S_{E,50}$ FOR PALOMARES PEOPLE



Mediana $\delta = 79.43$ mSv

Varianza $\sigma^2 = 0.31$

Media log. normal = 92.7 mSv

TABLE 1. REMEDIAL ACTIONS TAKEN AT PALOMARES
FOLLOWING THE JANUARY, 1966, ACCIDENT

Location	Hectares*	Acres	Remediation
Impact Point 2	1.6	4	Removed top 10 cm
Impact Point 3	0.6	1.5	Removed top 10 cm
Remainder	224	550	Plowed to 30 cm where possible
Total	226	558	

*259 hectares equal one square mile.

TABLE 2. DISTRIBUTION OF PEOPLE
IN RELATION WITH THEIR
PU-URINE ANALYSES.

Nº ANALYSES	PEOPLE		
	MEN	WOMEN	TOTAL
1	123	120	243
2	135	141	276
3	30	41	71
4	17	16	33
5	13	12	25
6	6	8	14
7	5	3	8
8	8	6	14
9	6	5	11
10	5	8	13
11	3	1	4
12	1	0	1
13	0	1	1

TABLE 3. DISTRIBUTION OF PEOPLE IN
RELATION WITH THE NUMBER OF
PU-URINE ANALYSES AND THEIR
POSITIVE RESULTS.

№ URINE ANALYSES \ № POSITIVE RESULTS						
	1	2	3	4	5	6
1	11					
2	10					
3	27	1				
4	24	2				
5	3	7			2	
6	3	5	3	1		
7		2	2		2	
8	5	2	2			
9	3	3	3	1	1	1
10	1	3	4	3		1
11		1	1	2		
12			1			
13		1				

TABLE 4. ESTIMATED COMMITTED EFFECTIVE DOSE EQUIVALENT BASED ON URINARY EXCRETION DATA AND ASSUMED ACUTE INHALATION

ESTIMATED DOSE mSv (rem)	NUMBER OF PEOPLE
< 20* (< 2)	659
20-50 (2-5)	22
50-100 (5-10)	22
100-150 (10-15)	6
150-200 (15-20)	5

* Assumed to be negative

MDA of radiochemical procedure is 0.37 mBq (~10 fCi) corresponding to ~18mSv (~1.8 rem)